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THE NATURE OF INDIVIDUAL RADIOACTIVE PARTICLES

I. SURFACE AND UNDERGROUND ABD PARTICLES FROM OPERATION JANGLE

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USNRDL-374

U.S. NAVAL RADIOLOGICAL DEFENSE LABORATORY

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USNRDL-374
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AW-7

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28 November 1952

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Thin sections of a large number of radioactive fall-out particles from Operation JANGLE were made. The thin sections were studied under a petrographic microscope and then radioautographed. It was found that the particles were composed almost wholly of glass with varying amounts of included mineral fragments and air bubbles. The radioactivity was distributed irregularly throughout the particles.

INTRODUCTION

The work covered by this report was undertaken to obtain a detailed description of the internal structure, mineralogical composition and distribution of radioactivity within the radioactive fall-out particles collected at Operation JANGLE.

Operation JANGLE reports show a difference of opinion existing among various investigators as to the nature of the radioactive fall-out particles.

Maxwell[1] reports that the "Particles appeared to be fused earth with adhering small metallic points and imbedded black specks. These points and specks varied from submicron size up to 15 microns, with the majority 0.5 to 2.0 microns in size. Observations made in the course of activity measurements indicated that the greater the number of these black specks the greater the activity".

Schorr and Gilfillan[2] in describing other investigators' work say in reference to the active fall-out particles, "Many of these exhibited the characteristics of a fused glass, sometimes throughout, but more often consisting of a dirt nucleus with a fused glass outer structure. The activity itself appears to be contained often in a small incrustation which was trapped in the outer layer of the fused glass".

Alexander, Blume, and Jefferson[3] report". . .most of the radioactivity in the fall-out material was concentrated in fused coatings on occasional mineral grains. . .". These authors do not give any observational evidence or references supporting their statement.

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[1] Roy D. Maxwell, Radiochemical Studies of Large Particles, Project 2.5a-3, Operation JANGLE, April 1952, 3.

[2] M. G. Schorr and E. S. Gilfillan, Predicted Scaling of Radiological Effects to Operational Weapons, Project 2.0, Operation JANGLE, June 1952, 57.

[3] L. T. Alexander, J. M. Blume, and M. E. Jefferson, Analysis of Test Site and Fall-out Material, Project 2.8, Operation JANGLE, March 1952, 16.

A more detailed description of the active fall-out particles was presented by Poppoff, Adams, and others.[4] Studying crushed particles under the petrographic microscope, these authors found that "The particles were composed exclusively of glass with varying amounts of included mineral fragments".

From these four papers, it is apparent that opinion is divided as to whether the active particles are composed of mineral or soil grains with a fused coating or whether they are glass throughout. As to the distribution of radioactivity in the particle, two of the papers described the activity as located principally in the outer shell; however, no experimental evidence was given to sustain this point of view. The other two papers did not report on the distribution of radioactivity.

The experimental work reported in this paper adequately resolves the conflicting views on the composition of the active fall-out particles and describes the distribution of radioactive material within the particles.

LABORATORY METHODS

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The method used for studying the internal composition and structure of fall-out particles was to make thin sections through the center of particles and to study the sections under the petrographic microscope.

Approximately one hundred active particles ranging in size from 0.5 to 10.0 mm in diameter were selected from the fall-out collected over a large area at Operation JANGLE. The active particles were selected by checking a great number of the fall-out particles of varying size and appearance with an end window GM tube. Those which showed a measurable amount of activity, about 0.02 mr/hr or more, were retained and the rest discarded.

These active particles were then mounted in plastic.[5] Molds were prepared by drilling cylindrical cavities in paraffin blocks. The holes

[4] I. G. Poppoff, et al., Fall-out Particle Studies, Project 2.5a-2, Operation JANGLE, April 1952, 40.

[5] The plastic used was Astrolite No. R-250, manufactured by Industrial Plastics Service, Oakland, California.

were 12 mm in diameter and about 10 mm deep. One or more particles were placed in each hole and liquid plastic poured in to a depth of about 5 mm. The plastic, to which a catalyst had previously been added, set in a few hours, and was completely hardened by baking at 60°C for an hour. The fall-out particles which had been on the bottoms of the molds were thus embedded securely in the bottoms of the plastic cylinders.

The bottoms of the plastic cylinders were ground by hand on a wet glass plate with No. 400 carborundum until approximately one-half of the diameters of the included particles had been ground away. The coarseness of the ground surface was diminished by grinding it for a few minutes with successively finer grades of carborundum and emery, and finally finishing with No. 1600 emery.

The ground surface was then permanently mounted on a glass microscope slide with Canada balsam, and the upper part of the plastic cylinder ground away on an electrically-driven abrasive wheel until the section remaining on the slide was about 0.25 mm thick. The section was then ground by hand as previously described until the correct final thickness was reached.

It was decided to use the standard petrographic thin-section thickness of 30 microns for these sections. A standard procedure for determining the correct thickness of petrographic thin sections is to grind until quartz grains within the section show a first-order gray interference color under the crossed nicols of a petrographic microscope. To enable this method to be used, small grains of quartz were included in each mold. The grinding of the sections was continued until the included quartz grains showed the characteristic first-order grayness under crossed nicols.

A drop of Shillaber's immersion oil and a cover glass were put on each completed thin section before they were studied under the petrographic microscope.

After completion of the petrographic examination, the cover glass was removed and the oil carefully wiped off. A piece of Eastman NTB stripping film, emulsion side out, was cemented over each thin section with a gelatin-alum cement. These units were placed in a light-tight box for three weeks, after which they were processed through developer. Each developed film was removed from its thin section and mounted on the microscopic slide next to the section. Comparative examination of the section and radioautograph revealed the distribution of radioactivity within the particles.

A petrographic study of the thin sections of the fall-out particles revealed that almost all the particles were composed throughout of a transparent and colorless (in thin section) glass.[6] Included in the glass were fragments of mineral grains and air bubbles. In most of the particles, the mineral grains constituted about 2 to 10 per cent and the bubbles 5 to 40 per cent by volume of the particle (see Figs. 1, 2, 5, 7, and 8). Some particles were found which had neither included soil (mineral) grains nor bubbles. In two cases active particles were found which consisted of a pebble of volcanic rock with a few blobs of glass adhering to the surface (see Fig. 3).

Most of the included mineral fragments were too small to be readily identifiable with the petrographic microscope. However, some of the grains were large enough and properly oriented to obtain an optic axis figure, in which instances they were identified as quartz or feldspar. All the included grains showed the characteristic first order gray color of quartz and feldspar under crossed nicols. Since about 75 per cent of the soil surrounding the JANGLE site consists of quartz and feldspar,[7] it seems probable that most of the included mineral fragments within the glassy fall-out particles should also be quartz and feldspar.

Nearly all the included mineral grains had an angular shape and were smaller than the quartz and feldspar grains found in the unaltered soil. The grains had the definite appearance of being shattered and subsequently embedded in the molten glass. In only three cases out of several hundred were there any signs of partial melting of the included mineral crystal (see Fig. 4). The great majority of the fragments exhibited sharp edges and were apparently unaffected by the surrounding glass.

Usually the mineral fragments were scattered at random throughout the glass. In a few cases they were more or less concentrated in a zone near the surface of the glass particle (see Fig. 5).

Many of the glass particles contained areas of glass which differed distinctly in texture and included mineral grains (see Fig. 4). Evidently

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[6] Glass means an amorphous substance, usually transparent, consisting ordinarily of a mixture of silicates.

[7] I. G. Poppoff, et al., op. cit., 38.

the particles had grown by accretion of previously solidified glass particles upon other, still molten, glass particles.

Many of the particles had been broken subsequent to solidification.

There was no observable evidence that the particles had formed either by the partial melting of single mineral grains or by condensation of vapor upon a mineral grain.

The general features of the fall-out particles, namely glass throughout with included soil (mineral) fragments and bubbles, were the same for particles from both the Surface (S) and Underground (U) Shots. However, a fairly large proportion of the S-shot particles were spherical or spheroidal (see Figs. 4, 5, and 6). These spheres were quite common in the small sizes around 0.5 mm in diameter. These spheres usually contained less included mineral crystals and air bubbles than did the irregular shaped particles. Some of the spheres were completely glass with no inclusions of any kind (see Figs. 7 and 8). Three spherical particles were found which consisted wholly of a pattern of roughly circular crypto-crystalline areas having the appearance of a devitrified glass.

Most of the S-shot fall-out particles in the larger sizes, 2.0 to 3.0 mm in diameter and greater, were irregular in shape and opaque in appearance due to the many included bubbles.

None of the fall-out particles from U-shot were spheres. However, active glass spheres from U-shot fall-out have been found in particle sizes smaller than those studied here.[8]

The radioactive particles from U-shot were all irregular in shape and opaque with a light brown color. Superficially they resembled the mineral grains with which they were found. In thin section they exhibited the typical transparent, colorless glassy structure with included mineral fragments and bubbles (see Figs. 7 and 8). A few U-shot particles were found which consisted of a black glass which was opaque even in thin section (see Fig. 9). In general these opaque particles seemed to have a higher specific activity than the transparent particles. The number of inclusions in the U-shot particles was usually much greater than in the S-shot particles. The large number of bubbles was exceptionally striking. Flow lines were often observed in the glass. The particles have the appearance of having been formed under rather violent conditions when large amounts of gas and shattered soil (mineral) grains were entrapped in a frothy mass of molten glass.

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[8] Unpublished work by the authors of this paper.

DESCRIPTION OF THE RADIOAUTOGRAPHS — USNRDL-374

The radioautographs of the thin sections revealed that the activity is distributed throughout the particles and not located exclusively on or near the surfaces.

The distribution of radioactivity in most of the particles is heterogeneous. The activity sometimes follows flow lines in the glass, but there is no systematic variation of activity within the particles. Adjacent areas of glass which show no visible discontinuity vary considerably in content of radioactive material.

The radioactivity seems to be associated exclusively with the glass phase of the particles. No radioactive mineral fragments were observed. Some of the glass itself is nonradioactive.

The distribution of activity within the small clear spheres from S-shot is fairly homogeneous, but the variation of specific activity from sphere to sphere is quite noticeable (see Fig. 6).

GENESIS OF THE FALL-OUT PARTICLES — USNRDL-374

The evidence presented above clearly rules out the possibility that the radioactive fall-out particles were formed either by the partial or complete melting of individual mineral grains or by the deposition of radioactive material on the surface of mineral grains.

The frothy, contorted appearance of a typical active particle suggests that the particles were formed by the violent mixing of molten earth, shattered mineral grains and radioactive fission products with the force of the explosion scattering the molten mass into the air.

Most of the particles have the definite appearance of having been formed from a melt and not from the condensation of vaporous material. The temperature of the melt was usually not sufficiently high to melt the included mineral fragments. However, examination of electron micrographs of the JANGLE fall-out made last year at this Laboratory revealed small spheres on the order of 0.1 micron in diameter. It seems probable that spheres of this size were formed by condensation of vapor. To what size spheres formed by this method could have grown under the conditions prevailing in an atomic bomb burst cannot easily be estimated. It does not seem impossible that the clear glass spheres found up to 0.5 mm in diameter could have originated in this manner.

Approved by:

E. R. Tompkins

E. R. Tompkins, Head
Chemical Technology Division

For the Scientific Director

Acknowledgment

The authors wish to thank P. D. LaRiviere for his generous cooperation in the preparation of the photomicrographs.

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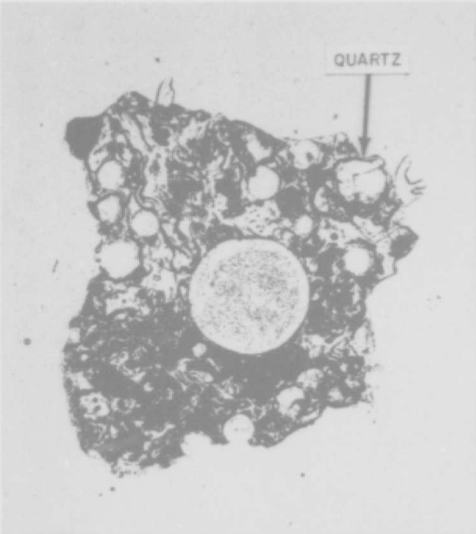
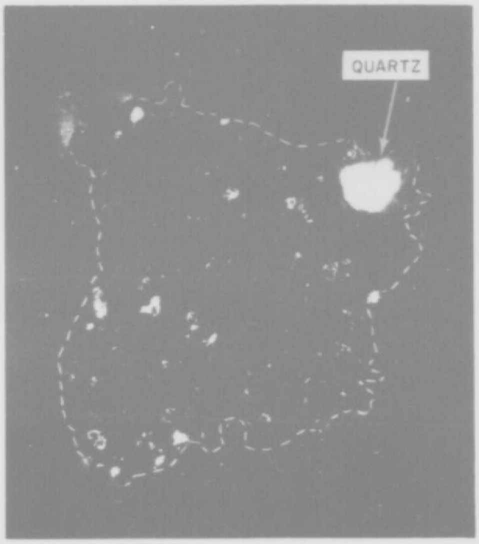


1a



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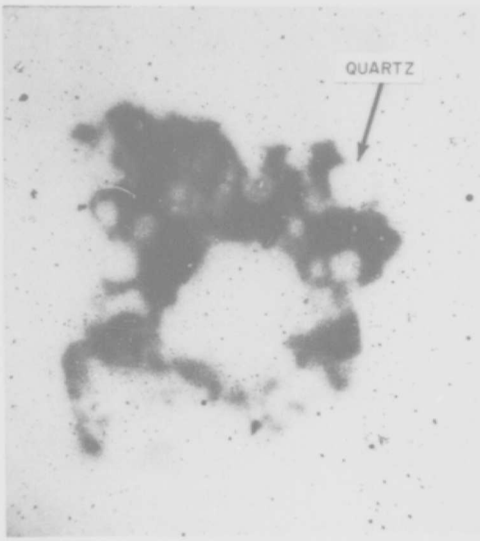

1c

FIG. 1 Scale : |—— 1 mm ——|

1a. Typical S-shot Fall-out Particle Photographed with Ordinary Light. The light, circular areas are air bubbles and the dark areas are froth and aggregates of small bubbles which do not transmit light. Notice the wavy, contorted appearance of the glass.

1b. The Same Particle Photographed with Polarized Light under Crossed Nicols. The glass does not transmit polarized light and appears black. The crystalline mineral grains which transmit the polarized light appear white. Notice the random distribution of the mineral fragments throughout the particle.

1c. Radioautograph of the Same Particle. The radioactivity is distributed irregularly throughout the particle. Notice that the quartz crystal in the upper right corner of the particle is non-radioactive.

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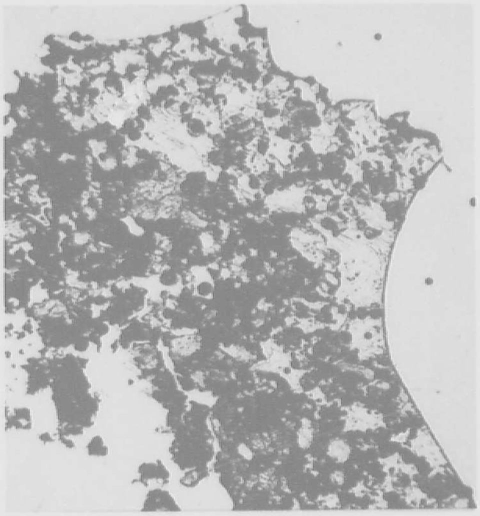
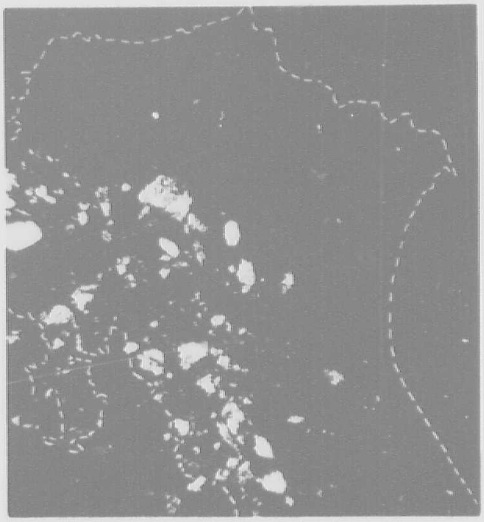


2a



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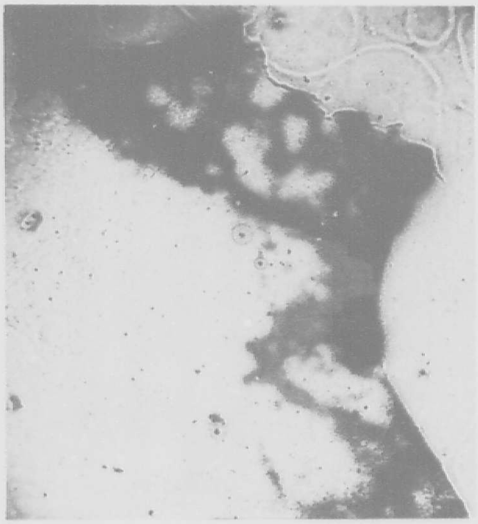


2c

FIG. 2 Scale ⊢ 1 mm ⊣

2a. S-shot Particle Photographed in Ordinary Light.

2b. Same Particle under Crossed Nicols. The mineral fragments are embedded in a glassy matrix occupying the lower left half of the particle.

2c. Radioautograph of the Same Particle. Notice that the radioactivity is associated with the glassy area and not with the crystalline area of the particle.

[redacted]

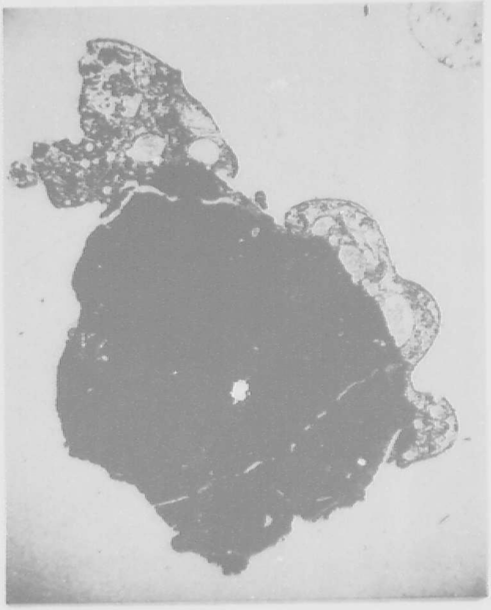


FIG. 3 Scale : ⊢ 1 mm ⊣

A Large Pebble of Volcanic Rock with Blobs of Radioactive Glass Adhering to Its Outer Surface. S-shot. Photographed with ordinary light.

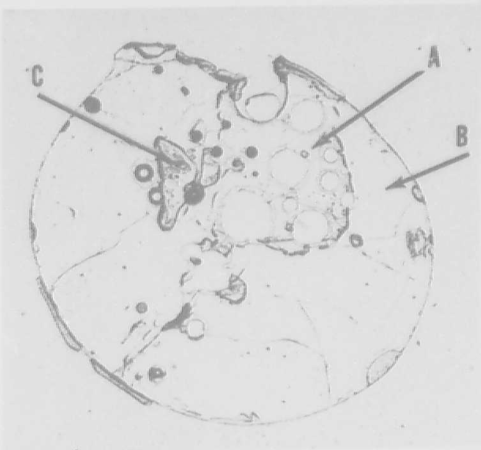


FIG. 4 Scale : ⊢ 1 mm ⊣

A Glass Sphere from S-shot Photographed with Ordinary Light Showing Two Distinct Areas of Glass (A and B). The glass fragment A apparently solidified first and then became embedded in fragment B while B was still molten. Also shown is a mineral fragment (C) which has almost completely melted in the glass. The partial melting is quite apparent when seen with polarized light, but does not show up well in ordinary light.

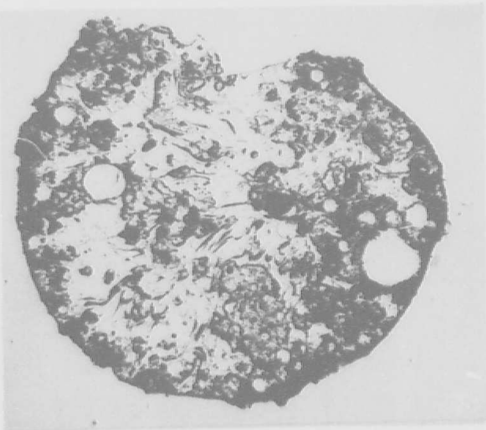
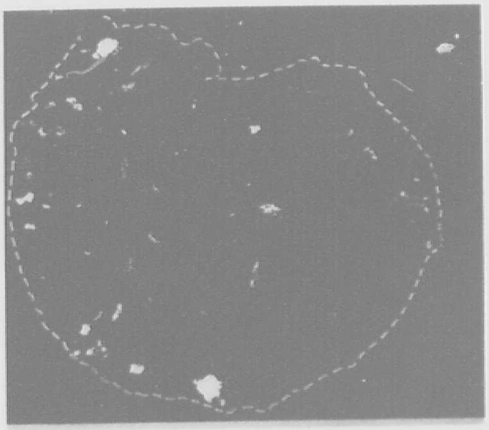


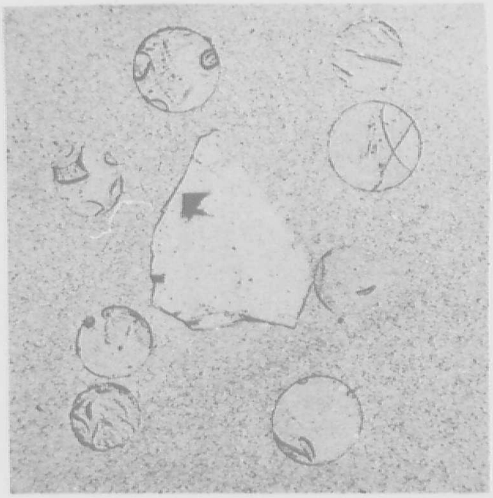
FIG. 5 Scale : ⊢ 1 mm ⊣

5a. A Typical Glassy, Spheroidal Particle from S-shot Photographed under Ordinary Light. Notice the many bubbles and flow lines in the glass.



5b. The Same Particle under Crossed Nicols. The included mineral fragments have a tendency to be concentrated near the outer edge.

[redacted]



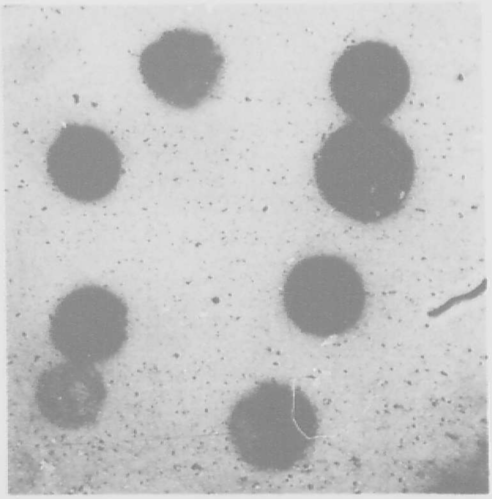
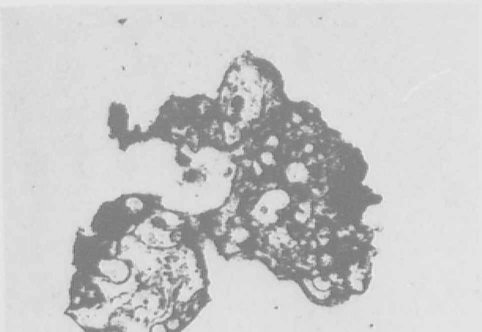


FIG. 6 Scale : ├── 1 mm ──┤

6a. Small Glass Spheres from S-shot Photographed with Ordinary Light. These spheres contain no included mineral grains or bubbles.

6b. Radioautographs of the Same Spheres. In most of the cases the activity is distributed homogeneously throughout the particles. However, notice the irregular distribution of activity in the upper left sphere. The specific activity of the glass apparently varies from sphere to sphere.



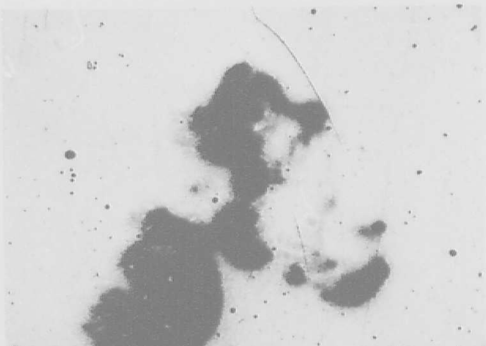


FIG. 7 Scale : ├── 1 mm ──┤

7a. Typical U-shot Particle Photographed with Ordinary Light. Notice the bubbles and flow lines. The dark areas are the frothy parts of the particle. The particle is completely glass.

7b. Radioautograph of the Same Particle Showing Heterogeneous Distribution of Radioactivity.

[redacted]

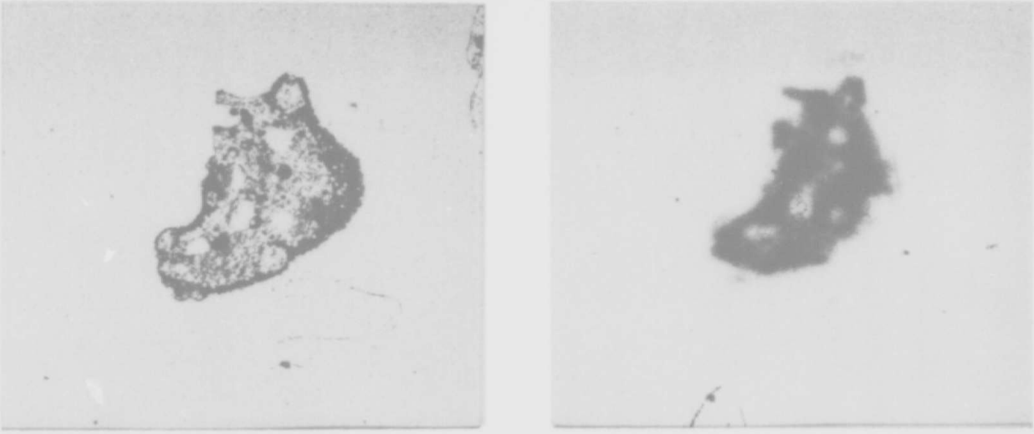


FIG. 8 Scale : ├──── 1 mm ────┤

8a. Typical U-shot Particle Photographed with Ordinary Light. The particle is completely glass.

8b. Radioautograph of the Same Particle Showing that the Activity is Fairly Evenly Distributed throughout the Particle. Most of the clear areas in the autograph correspond to air bubbles in the particle.

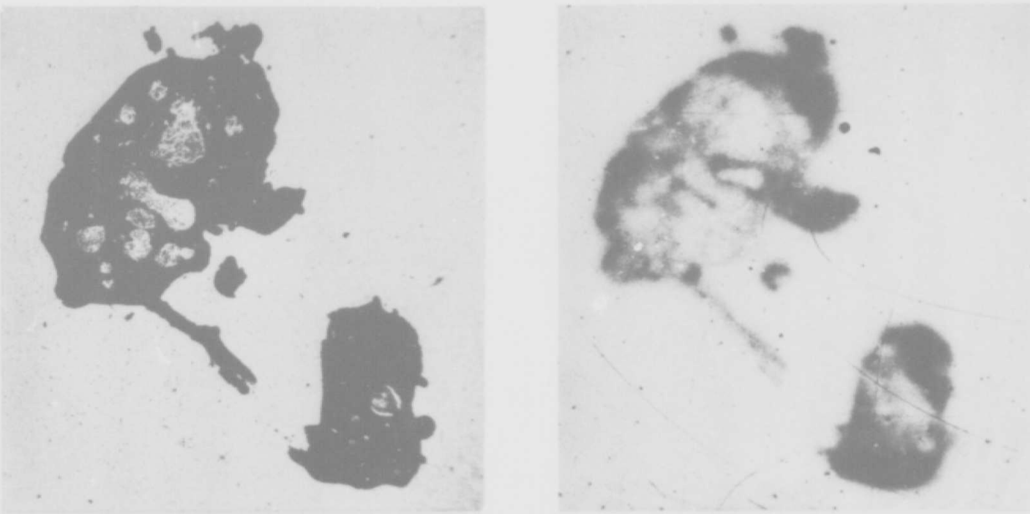


FIG 9 Scale : ├── 1 mm ──┤

9a. An Opaque, Glassy Particle from U-shot Photographed under Ordinary Light. Light areas are air bubbles. (The original single particle has been separated into two parts by an air channel running through it in the plane of the thin section.)

9b. Radioautograph of the Same Particle.